



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

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Applicant (s): Alex Kuperman et al.

Serial No.: 09/544,742

Group Art Unit: 1754

Filed: April 7, 2000

Examiner: Steven P. Griffin

For: METHOD OF PREPARING A CATALYST CONTAINING GOLD AND
TITANIUM

I HEREBY CERTIFY THAT THIS CORRESPONDENCE IS BEING DEPOSITED WITH THE UNITED STATES POSTAL SERVICE, WITH SUFFICIENT POSTAGE, AS FIRST CLASS MAIL, IN AN ENVELOPE ADDRESSED TO: COMMISSIONER FOR PATENTS, WASHINGTON, D.C. 20231,	
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Sir:

BRIEF FOR APPELLANT

Pertaining to the above-captioned patent application, and pursuant to a Notice of Appeal filed therein on April 2, 2002, submitted herein for entry into the record is a Brief for Appellant, which constitutes an Appeal from a Final Rejection, dated December 10, 2001, of Claims 1-32.

REAL PARTY OF INTEREST

The real party of interest in this Appeal is The Dow Chemical Company.

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RELATED APPEALS AND INTERFERENCES

No Appeals are pending related to the instant Appeal.

The following potential interference is noted for the record. The claims to US Patent Application Serial No. 09/209,698 have been held to be allowable pending a potential interference with US 5,932,750. As of the current date, Appellant is not aware that an interference has been declared. Both US Patent Application Serial No. 09/209,698 and US 5,932,750 contain claims related to a catalyst composition comprising gold on titania, its use in a hydro-oxidation process, as well as claims to a method of making the catalyst composition (See Claims 98 and 99 in application Serial no. 09/209,698). Appellant sees patentable differences between the claims under this Appeal and the claims in the potential interference and will address any questions to this point as raised by the Board.

STATUS OF CLAIMS

Claims 1-32 are pending in the application and are the subject of this Appeal.

Claim 33 was canceled by Appellant and is not the subject of this Appeal.

Claims 34-36 were canceled as non-elected claims, in response to a restriction requirement. Claims 34-36 are not the subject of this Appeal.

STATUS OF AMENDMENTS

Claims 1, 2, 3, 12, 14, 17, 18, 19, 20, 23, 24, 25, and 30 were amended primarily with minor amendments to clarify the language of the claims. The amendments were entered by the Examiner.

Claims 33-36 have been canceled in the manner noted hereinabove. The Examiner has entered the cancellation of Claims 33-36.

SUMMARY OF THE INVENTION

Claims 1-32 pertain to a process of preparing a catalyst composition comprising gold on a titanium-containing support, the process comprising impregnating a gold compound and a reducing agent onto a catalyst support, wherein the reducing agent comprises titanium, or the catalyst support comprises titanium, or both the reducing agent and catalyst support comprise titanium, under conditions sufficient to prepare the catalyst composition.

The catalyst composition prepared by the claimed process is useful in hydro-oxidation processes wherein, for example, an olefin, such as propylene, is oxidized with oxygen in the presence of hydrogen and the catalyst to prepare an olefin oxide, such as propylene oxide. The advantages of the claimed catalyst preparation method are discussed in detail in the arguments hereinbelow.

ISSUES

Should the Final Rejection of Claims 1-9, 18, 20, 22-27, and 29-32 under 35 U.S.C. 102 (b) for allegedly being anticipated by US 5,051,394 (hereinafter Haruta '394) be maintained?

Should the Final Rejection of Claims 11-14, 16-17, 19, 21, and 28 under 35 U.S.C. 103(a) for allegedly being unpatentable over US 5,051,394 (Haruta '394) be maintained?

Should the Final Rejection of Claims 10-17 under 35 U.S.C. 103(a) for allegedly being unpatentable over Haruta '394, further in view of US 5,532,030 (hereinafter Hirose et al.) be maintained?

Is reference EP-A1-0,709,360 (Haruta 'EP) a material reference? Does it present evidence teaching away from impregnation, which would lend unobviousness to Appellant's claimed impregnation process?

GROUPING OF CLAIMS

Claims 1-9, 18, 20, 22-27, and 29-32 form Group A; but the claims do not stand or fall together. Claims 1-9, 18, 25, 26, and 29-32 shall be considered together as Subgroup A¹. Claim 20 (Subgroup A²) shall be argued as an independent subgroup. Claims 22, 23, 24, and 27 (Subgroup A³) shall also be argued independently of the other subgroups.

Claims 11-14, 16-17, 19, 21, and 28 form Group B; but these claims do not stand or fall together. Claims 13, 14, 16, 19, and 21 shall be considered as Subgroup B¹. (Claim 10 shall be considered in this group; see remarks below.) Claims 11 and 12 shall be considered as Subgroup B². Claim 17 shall be considered separately as Subgroup B³, and Claim 28 shall be considered separately as Subgroup B⁴.

Claims 10-17 form Group C; but these claims do not stand or fall together. Claims 10 and 13-16 shall form Subgroup C¹. Claims 11 and 12 shall form Subgroup C². Claim 17 shall form Subgroup C³.

ARGUMENTS

A. Concerning the Final Rejection of Group A: Claims 1-9, 18, 20, 22-27, and 29-32 Under 35 USC 102(b)

Claims 1-9, 18, 20, 22-27, and 29-32 stand finally rejected under 35 USC 102 (b) as allegedly being anticipated by US 5,051,394 (Haruta '394) for the reasons of record. This Final Rejection is appealed for the following reasons.

A.1. Summary of the Invention

The instant application pertains to a process of preparing a catalyst composition comprising gold on a titanium-containing support, the process comprising impregnating a gold compound and a reducing agent onto a catalyst support. The titanium is required to be derived from the reducing agent, or the catalyst support, or both the reducing agent and the catalyst support. As a first example, titanium may be derived from the support, e.g.: (1) A gold compound and an

organic reducing agent (e.g., carboxylic acid or an alcohol) are impregnated onto a titanium-containing support, (e.g., titanium oxide or titanosilicate). As a second example, titanium may be derived from the reducing agent, e.g.: (2) A gold compound and an organotitanium compound or a titanium coordination compound are impregnated onto a support without titanium (e.g., silica). As a third example, the titanium may be derived from both the reducing agent and the support, e.g.: (3) A gold compound and a reducing agent containing titanium, such as an organotitanium compound or titanium coordination compound, are impregnated onto a catalyst support containing titanium (e.g., titanium oxide or titanosilicate).

A.2. Summary of Prior Art

Haruta '394 discloses a method of preparing a composition comprising gold particles immobilized on a metal oxide, for example, titanium oxide. The reference summarizes various precipitation methods of preparing catalysts: (1) coprecipitation method, (2) uniform deposition and precipitation method, (3) dropwise neutralization and precipitation method, (4) reductant addition method, and (5) pH-controlled neutralization and precipitation method. (Haruta '394, column 1, line 18, continuing all the way to Column 2, line 57) In the co-precipitation method taught at Columns 3 and following, an aqueous solution containing a gold compound and a water-soluble metal salt, e.g., titanium salt, is neutralized with an alkali aqueous solution to yield a coprecipitate (essentially a gold hydroxide and metal hydroxide co-precipitate) that coprecipitates from the solution. A carboxylic acid is added prior to or after formation of the coprecipitate. The coprecipitate is aged, separated, and heated at a temperature in the range from 100°C to 800°C to obtain a composition comprising reduced gold particles immobilized on an oxide (e.g., titanium oxide). (Haruta '394, Column 3, lines 14-55; Column 4, lines 25-45; Column 7, lines 9-17)

A.3. Arguments in Favor of Novelty of Subgroup A1:

Claims 1-9, 18, 20, 25, 26, and 29-32

In a rejection under 35 U.S.C. §102 (b), the cited reference must disclose explicitly or implicitly each of the elements that Applicants claim. *Stryker Corporation v. Davol, Inc.*, 57 USPQ 2d 1133; 234 F.3d 1252 (Fed. Cir. 2000);

Scripps Clinic & Research Foundation v. Genentech, Inc., 18 USPQ 2d 1001, 1010; 927 F.2d 1565, 1576 (Fed. Cir. 1991).

Haruta '394 *taken in its entirety* relates to precipitation and coprecipitation methods for preparing a gold-titanium oxide composition. (Co)precipitation is an "ages-old" chemical technique wherein a solution containing a soluble compound of the desired element is treated with a precipitation agent to cause the formation of a less soluble compound (salt or oxide/hydroxide) of the desired element. When the concentration of the less soluble compound exceeds its solubility limit in the solution, the less soluble compound settles out of, "precipitates from," the solution as particles. In Haruta '394, an aqueous solution containing a water-soluble gold compound, a water-soluble metal salt (precursor to metal oxide, e.g., titanium oxide), and a carboxylic acid is treated with an alkali hydroxide at controlled pH to cause the hydroxides/oxides of the gold and metal to coprecipitate. Then, in the presence of the carboxylic acid, the precipitate is heated to cause the gold oxide/hydroxide to be converted to ultra-fine metallic gold particles on a titanium oxide support. Significantly, Haruta '394 is silent with respect to an impregnation method of preparing the catalyst.

The (co)precipitation method of Haruta '394 disadvantageously requires large amounts of solvent that ultimately must be recovered for reuse, else the method is impractical. As a further disadvantage, Haruta's method requires that pH be controlled throughout the precipitation process, so as to obtain gold of the correct particle size. (Haruta '394, Column 5, lines 27) Controlling pH adds a complexity to the method, which is then not amenable to large-scale operation. More disadvantageously, the (co)precipitation method must be conducted slowly to ensure precipitation of small gold particle size and proper adherence of gold to the support. As an even further disadvantage, the disclosed (co)precipitation method does not provide for reproducible control over the amount of gold deposited onto the support. From batch to batch, differing amounts of gold precipitate and adhere to the support. Finally, any gold remaining in solution or precipitating into the solution rather than on the support must be recovered, else the costly gold metal is wasted.

In contrast, the method of the invention requires an impregnation technique, which differs significantly from the (co)precipitation technique of Haruta '394. Appellant's impregnation technique involves preparing an impregnation solution containing a soluble gold compound, and likewise the same or a different solution containing a soluble compound of reducing agent, and then simply *applying* the solution(s) (e.g., *wetting*) to a catalyst support. In a preferred embodiment, the impregnation involves wetting the catalyst support only to the point of "incipient wetness." Then, the wetted support is dried so as to evaporate the solvent and leave the desired elements adhered to the support.

The claimed impregnation method provides many advantages over the precipitation methods of Haruta '394. Specifically, the claimed impregnation method requires less quantity of solvent as compared with Haruta's precipitation method. Only the amount of solvent to be applied to the support is needed. In the impregnation method, there is no need to recover large amounts of excess solvent for reuse. The claimed impregnation method may also beneficially employ non-aqueous solvent; there is no absolute requirement to use water, as is the case with Haruta '394. The claimed impregnation method does not require the complexity of controlling pH throughout the deposition process. As a further advantage, the claimed impregnation method can be conducted more quickly than the precipitation method of the prior art, which requires a longer time for small particle precipitation. As a further advantage, the claimed impregnation method provides for better control over the amount of gold deposited onto the support. Since a measured amount of gold is applied directly to the support, the quantity of gold deposited is acceptably consistent from batch to batch. Most advantageously, the claimed impregnation method does not require recovery of unused gold from the solution, because no unacceptable excess of gold solution is employed and the solution used is directly applied to the support. Clearly, the claimed impregnation method is significantly more favorable for commercial, large-scale preparations, as compared with the prior art method.

The Examiner is not persuaded that the term "impregnation" is patentably distinguished over "precipitation," insisting that "impregnation is known in the art to involve precipitation and the two methods are not recognized in the art as mutually

exclusive... ." To this point, the Examiner misunderstands the techniques and therefore errs in his conclusion. In fact, the terms "impregnation" and "precipitation" are recognized in the art to be distinguishable over each other. Several publications in the field of catalysis, of record in this application, provide evidence of the strength of Appellant's position. The Board's attention is drawn, for example, to the following quotation from a well-known textbook in heterogeneous catalysis:

"Either of two types of processes, generally termed the *precipitation method* and the *impregnation method*, is commonly used for making catalysts."

Heterogeneous Catalysis in Practice, Charles N. Satterfield, Professor of Chemical Engineering MIT, McGraw-Hill Book Company, New York, 1980, pp. 70.
(Copy of record; italics in original.)

Satterfield also teaches at page 70 that the precipitation method involves "in the initial stages the mixing of two or more solutions or suspensions of material, causing precipitation;..." Details on precipitation are presented at page 72 of Satterfield. Again at page 70, last paragraph, Satterfield teaches, "If a carrier is to be incorporated in the final catalyst, the original precipitation is usually carried out in the presence of a suspension of the finely divided support" This latter method is typically referred to as "deposition precipitation." In contrast, referring to impregnation, Satterfield teaches:

"*Impregnation* is the easiest method of making a catalyst. A carrier, usually porous, is contacted with a solution, usually aqueous, of one of more suitable metallic compounds. The carrier is then dried, ..."

Heterogeneous Catalysis in Practice, Ibid., p. 71.
(Copy of record; italics in original.)

At pages 82 and 83, Satterfield gives details on impregnation, including the "dipping method" and the "impregnation to incipient wetness" ("dry impregnation") method.

Other artisans skilled in the field also recognize the differences between impregnation and precipitation. See, for example, the following citations also of record in the application:

- ** Table XIII, wherein the preparation method of preparing a sulfur-free Ni/SiO₂ catalyst is disclosed as either:
 "Various methods of coprecipitation at pH 8.5,"
 "Dry mix,"
 "Impregnation at 75°C"
 "Structure and Activity of Silica-Supported Nickel Catalysts,"
 J. W. E. Coenen and B. G. Linsen, in *Physical and Chemical Aspects of Adsorbents and Catalysts*, B. G. Linsen, editor, Academic Press, New York, N.Y., 1970, p.501.
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- ** "The active precursor can be applied onto the support by:
 -adsorption
 -impregnation and drying
 -precipitation".

J. W. Geus, "Production and Thermal Pretreatment of Supported Catalysts," in *Preparation of Catalysts III*, G. Poncelet, P. Grange, and P. A. Jacobs, editors, Elsevier Science Publishers B.V., Amsterdam, 1983, p. 6.

The latter reference distinguishes impregnation on pages 6-7 from precipitation/deposition-precipitation on page 8. (Geus, *ibid*, pp. 6-8, of record). In view of the above, Appellant fairly asserts that impregnation and precipitation are recognized as different techniques in the art of catalyst preparation.

The Examiner asserts that two references: US 5,571,771 (Abel et al.) and US 5,700,753 (Wang et al.) illustrate the preparation of an impregnated catalyst made by precipitation; and thus the two techniques are not mutually exclusive. The Examiner misunderstand the technical details of these references and therefore errs. Abel et al. teaches the impregnation of a salt (e.g., gold salt) onto a support by contacting a solution of the salt with the support. Abel et al. teaches further that when an *impregnated* salt is too soluble or contains poisonous anionic species (e.g., chlorides), the impregnated salt may thereafter be "converted to an insoluble form by precipitation with hydroxides." (Abel et al., Column 3, lines 8-17) The referred to "precipitation" actually comprises a surface conversion of an already-impregnated, but too soluble salt into an impregnated, insoluble hydroxide. The overall process involves two steps: impregnation to place the salt onto the surface of the substrate,

followed by precipitation with base (fixation as an insoluble hydroxide). The impregnation step itself does *not* involve precipitation. Notably, in describing the *impregnation step*, Abel et al. teaches away from precipitation, "The solution of the salts should be at a temperature which is high enough *to prevent precipitation of the salts during application to the support*." (Abel et al., Column 4, lines 21-23; emphasis added.)

The Examiner refers to the Table at Column 8 of Abel et al. as teaching impregnation with precipitation (Table footnote referring to Comparative Example 3b: "#Surface impregnated catalyst in accordance with EP-A-0 519 435 (precipitation with base)". Comparative Example 3b (Abel, Column 6) teaches that a solution of sodium chloropalladate was applied to a support with an atomizer (an obvious impregnation technique), and then the impregnated support was dried in a hot air stream. *Thereafter*, the impregnated support was treated with sodium hydroxide so as to precipitate palladium and gold hydroxides (i.e., fix an insoluble form onto the support). Note that Abel et al. at this example teaches three distinct steps: impregnation, drying, and thereafter precipitation.

Likewise, Wang et al. describes a two-step method of preparing a catalyst, which involves first impregnation and *thereafter* fixation with a basic alkali metal salt (referred to as precipitation) to secure the impregnated salt to the support. (Wang et al, at Column 2, lines 30-37) Again, the impregnation step is followed by a distinct precipitation step to form an insoluble and fixated form of the salt.

In view of all of the above, it is submitted that Abel et al. and Wang et al. disclose *multi-step* catalyst preparation processes, wherein impregnation is followed by precipitation. The two steps do not occur simultaneously and therefore are independent of each other.

The Examiner maintains that "Haruta '394 discloses gold particles immobilized on a titanium dioxide carrier ..., which would inherently entail gold particles impregnated onto the carrier." To the contrary, the word "immobilized" *by itself* does not inherently imply *any specific* method of preparation; it is simply a broad description. Significantly, Haruta '394 teaches in clear details the precipitation

and coprecipitation methods, while making no mention of details characteristic of impregnation. Accordingly, Haruta '394 cannot be interpreted to include inherent teachings to impregnation.

The Examiner maintains that "in any case, Applicant appears to admit that 'impregnation' is known in the art and therefore cannot, by itself, be the basis for patentability." The Examiner's argument lacks substance. Appellant admits that impregnation is known in the art, as is crystallization, fractional distillation, precipitation, and various other chemical processing techniques. That general knowledge does *not*, in itself, preclude obtention of a patent in all of these areas of chemical processing. Patentability lies in the details of the claims "*taken as a whole.*" Appellant seeks a patent for a method characterized by specific elements combined with a specific process step for preparing a specific catalyst (*impregnation of a gold compound and a reducing agent onto a support; the reducing agent, or the support, or both reducing agent and support comprising titanium, the impregnation being conducted under conditions sufficient to prepare a catalyst comprising gold on a titanium-containing support*). The specific limitations render Appellant's impregnation process patentable.

The Final Rejection states that the "features upon which the Applicant relies ... are not recited in claim 1," such features including the following (as compared with the precipitation method of Haruta '394): (1) Lower amounts of solvent can be employed. (2) The deposited amount of gold is more reproducible from batch to batch. (3) There is no need to control pH. (4) The time for depositing metal onto the support is quicker. These advantages are *inherent characteristics* of the claimed impregnation technique and are *inherently distinguishable* over Haruta '394's precipitation technique (which implicitly would be recognized by one skilled in the art to involve larger amounts of solvent, less reproducibility, a need to control pH, and longer deposition times). Refer to the Satterfield reference (*ibid.*), which describes the impregnation method as the "easiest method for preparing catalysts." Accordingly, Appellant is not required to add inherent features to the claims, especially such features that might vary among different embodiments of the invention depending upon the specific conditions employed. The limitations that

characterize the claimed process and overcome Haruta '394 are already present in the claims.

The Final Rejection again argues that features upon which Applicants rely, in this instance "wetting to the point of incipient wetness," are not recited in Claim 1. Appellant sees no reason to limit Claim 1 to this particular preferred embodiment. Haruta '394 is silent with respect to the limitations of Claim 1 "*taken as a whole*;" therefore, Claim 1 is patentable without the additional limitation to "incipient wetness."

At page 9 the Final Rejection emphasizes a second time that "impregnation is known in the art to involve precipitation and the two methods are not recognized in the art as mutually exclusive." The Examiner states, "Claim 1 recites only impregnation and Haruta '394 discloses loading and immobilization onto the support." Appellant vigorously disagrees. The terms "loading" and "immobilization," taken by themselves, are so general as to be non-descriptive as to the method involved. Taken into account its complete disclosure, Haruta '394 unquestionably relates to deposition-precipitation and co-precipitation techniques for catalyst preparation. Impregnation is clearly a different technique, as evidenced by the authoritative documents of record in this application. Appellant is claiming a different method of catalyst preparation (not the catalyst composition, by the way).

The rejection cites a list of specific locations in Haruta '394 that allegedly disclose the claimed subject technology of Claim 2 (specific gold compounds), Claim 3 (atomic ratio of Au/Ti), Claim 4 (reduction with carboxylic acid), Claims 5-7 (carboxylic acids and salts thereof), Claim 8 (at least 1 mol per mol), Claim 9 (citric acid with coprecipitate), Claim 18 (titanium oxide carrier), Claim 20 (0.05 mol titanium sulfate and atomic ratio of Au/Ti), Claims 22-23 (alkali compound to metal salt), Claim 24 (use of sodium carbonate), Claim 25 (magnesium citrate), Claim 26 thorough washing), Claim 27 (use of sodium carbonate), Claim 29 (20-90 deg C), and Claims 30-32 (drying and firing in air at 400 deg C). Without delving into each of these alleged teachings, Appellant vigorously maintains that none of the citations disclose the claimed invention, because all of Claims 2, 3, 4, 5-7, 8, 9, 18, 20, 22-23,

24, 25, 26, 27, 29, and 30-32 depend from Claim 1, which requires impregnation, which is a significantly different technique from the (co)precipitation technique of Haruta '394. Accordingly, none of Appellant's claims is anticipated by Haruta '394.

A.4. Additional Argument in Favor of Novelty of Subgroup A²: Claim 20

In addition to the above, Claim 20 (Subgroup A²), which requires a titanium loading on the support of greater than 0.02 weight percent and less than 20 weight percent, is further removed from the anticipation rejection. Haruta '394 is silent with respect to an explicit disclosure of titanium loading. Implicitly, Haruta '394 *may* disclose a titanium loading by virtue of disclosing TiO₂ and mixtures of Au/TiO₂ wherein the Au/Ti atomic ratio is 1/19. From these disclosures, the titanium loading may be calculated as shown in Appendix B, attached hereto. Thus, implicitly, the Ti loading in TiO₂ is 59.1 weight percent, while the Ti loading in mixtures of Au/TiO₂ having a Au/Ti atomic ratio of 1/19 is 53.1 weight percent. Significantly, these implicitly disclosed values lie outside the range claimed by Appellants in Claim 20 (>0.02 and <20 weight percent). Hence, Claim 20 is not anticipated by Haruta '394.

A.5. Additional Argument in Favor of Novelty of Subgroup A³: Claims 22, 23, 24, and 27

Claims 22, 23, 24, and 27 (Subgroup A³) require that one or more promoter metals, such as alkali compounds, be *impregnated* onto the support. The Examiner argues that Haruta '394 discloses adding an alkali compound that would promote precipitation by adjusting pH. Significantly, Haruta '394 makes no disclosure or suggestion that the alkali metal ion (e.g., sodium ion) in combination with the hydroxide or carbonate anion can function as anything other than a pH adjuster. In contrast, Appellant claims the alkali metal itself as a promoter to enhance the productivity of the catalyst. Whether the alkali compound promotes precipitation would depend on the specific alkali compound used. Some would promote precipitation; others would not. The claim requirement for impregnation would inherently demand the use of a promoter metal compound *that essentially does not cause precipitates*. Once precipitation begins, the deposition precipitation method would commence; the disadvantages thereof would control; and the deposition would

no longer be considered an impregnation. Accordingly, Claims 22, 23, 24, and 27 are not anticipated by Haruta '394.

Thus, in view of all of the above, it is submitted that all of Claims 1-9, 18, 20, 22-27, and 29-32 clearly meet the novelty requirements of 35 U.S.C. 102(b). It is therefore respectfully requested that the Final Rejection under 35 USC 102(b) be reversed.

B. Concerning the Rejection of Group B: Claims 11-14, 16-17, 19, 21, and 28 Under 35 USC 103(a) in view of Haruta '394

Claims 11-14, 16-17, 19, 21, and 28 stand finally rejected under 35 USC 103(a) as allegedly being unpatentable over Haruta '394, for the reasons of record. Haruta '394 is described in Section A hereinabove. From the Examiner's remarks, Appellant believes that Claim 10 may be intended to be included in this rejection. Accordingly, this Final Rejection, including as may pertain to Claim 10, is appealed for the following reasons.

B.1. Arguments in Favor of Unobviousness of Subgroup B¹: Claims 10, 13, 14, 16, 19, 21

A proper analysis under §103 requires, *inter alia*, consideration of whether the prior art would have motivated or suggested to those of ordinary skill in the art at the time the invention was made that they should modify the prior art along the lines of the claimed invention. *In re Gordon*, 733 F.2d 900, 221 USPQ 1125, 1127 (Fed. Cir. 1984); *In re Vaeck*, 947 F.2d 488, 493 20 USPQ2d 1438, 1443 (Fed. Cir. 1991).

With respect to Claim 10 ("reducing agent contains titanium"), the Examiner maintains that Haruta '394 suggests the use of reducing agents containing titanium. The Examiner reasons that because Haruta '394 discloses carboxylic acid reducing agents and titania supports, one skilled in the art would be motivated to select titanium for the reducing agent more than any other of the 68 metals of the Periodic Table. The Examiner errs in his reasoning.

First, it should be recognized that a reducing agent and a catalyst support function in distinctly different ways. A support is typically a relatively inert carrier of the catalytic metal, typically functioning to spread-out the catalytic metal over a relatively large surface area. In contrast, a reducing agent is capable of transferring electrons to a reducible atom or compound, herein gold ($\text{Au}^{+3} \rightarrow \text{Au}^{<+3}$). To recall from general chemistry, *during reduction, the reducing agent is itself oxidized*, since it is losing electrons. In consideration of this basic chemistry, the Examiner's assertion that one skilled in the art would be motivated to select a reducing agent containing titanium, because titania is mentioned as a support material by Haruta '394, is far-fetched. There simply is no technical nexus between a support and a reducing agent. Moreover, one skilled in the art would know that the titanium in titania (TiO_2) is already in a maximum oxidation state of +4 and cannot give up more electrons (act as a reducing agent) to form a higher oxidation state. (An oxidation state of greater than +4 is not known for titanium.) All the more reason why the mental connection of substituting titanium from the support as a reducing agent would not be made by one skilled in the art. Hence, the necessary motivation in the art is missing. Clearly, the Examiner impermissibly employs "hindsight" to construct an argument against the claim. Moreover, even if *arguendo* it were "obvious to try" titanium as the reducing agent, there is no indication or motivation in Haruta '394 that such a combination would be successful in the preparation of an operable gold-titanium catalyst. "Obvious to try" is not a valid means for defeating a claim.

With regard to Claims 11, 13, and 14 (which embrace various titanium carboxylates as reducing agents), the Examiner argues that a carboxylic salt "is considered to be an alkyl compound and a carboxylate compound;" and therefore, Haruta '394 by teaching carboxylate salts and titania supports would lead "one of ordinary skill to use titanium as a salt." On the contrary, Claims 11, 13 and 14 are unobvious, because Haruta '394 only mentions titanium in a non-material manner, that is, as a support. The support contains titanium in a maximum oxidation state and functions differently from a reducing agent. (*Vida supra.*) There is no motivation in Haruta '394 to introduce a component of the support into the reducing agent.

With regard to Claim 16 (gold compound deposited onto support prior to titanium-containing reducing agent), the Examiner again relies on an erroneous connection between precipitation and impregnation methods. Appellant has already shown these methods to be patentably distinct. (*Vide supra*)

The Examiner's reasoning for rejection Claims 19 and 21 are repetitious of the above arguments. Appellant maintains that these claims are also unobvious for the same reason, that there is no nexus between the function of a support and the function of a reducing agent and no motivation in the cited reference to make a connection between the two.

B.2. Additional Argument in Favor of Unobviousness of Subgroup B2: Claims 11 and 12

Claim 11 requires an organotitanium reducing agent, described in the specification at page 9, lines 2-7, as a compound comprising titanium bonded to a carbon atom by a σ -bond or a π -bond. Claim 12 requires reducing agent species of alkyltitanium or cyclopentadienyltitanium compounds, which inherently contain a Ti-C σ -bond or π -bond, respectively. In addition to the arguments of the above section, it is noted that carboxylate salts such as those disclosed by Haruta '394 are characterized by a metal ion being bonded to *the oxygen* of the carboxylate group. No *metal-carbon* bond is found in the carboxylates disclosed by Haruta '394. Conversely, carboxylate is neither present nor involved in the compounds of Claims 11 and 12. Accordingly, Claims 11 and 12 are clearly non-obvious, because Haruta '394 neither teaches, suggests, nor even hints at using the claimed σ - and π -bonded organotitanium compounds as reducing agents.

B.3. Additional Argument in Favor of Unobviousness of Subgroup B³: Claim 17

With regard to Claim 17 (Ti loading greater than 0.02 and less than 20 weight percent), the Examiner finds the claim obvious, based on Haruta '394's disclosure of a gold (Au) to titanium (Ti) atomic ratio of 1/19, which is "slightly above 5%." The Examiner errs by comparing apples with oranges: the disclosed gold/titanium atomic ratio of 1/19 versus Appellant's claimed Ti loading of greater

than 0.02 to less than 20 weight percent titanium. The comparison is invalid. Moreover, the Examiner incorrectly insists that "units of measurement are not considered to provide patentable weight and 1/19 is slightly above 5%." This argument totally lacks merit. Units of measurement define the parameter measured, and in so doing, can provide patentable weight.

Appellant maintains that Haruta '394 makes no explicit teaching to titanium loading, but may be considered to make two implicit teachings thereto. A titania support (TiO_2) is disclosed; so a skilled artisan might calculate the weight percentage of Ti in TiO_2 to be 59.9 weight percent. Likewise, a mixture of Au/ TiO_2 having an atomic ratio of Au/Ti of 1/19 is disclosed. A skilled artisan might calculate the weight percentage of Ti in that mixture to be 53.1 weight percent. (Refer to Appendix B attached.) These inherent values are well outside the claimed titanium loading (0.02-20 wt. percent), and there is no suggestion or hint in Haruta '394 to use a titanium loading within Appellant's' claimed range.

B.4. Additional Argument in Favor of Unobviousness of Subgroup B⁴: Claim 28

With regard to Claim 28 related to impregnation to the point of incipient wetness, the Examiner asserts that the Claim is obvious over Haruta '394, because impregnation and precipitation are not mutually exclusive. Appellant has submitted herewith strong evidence to the contrary. Moreover, the Examiner's argument makes no sense. How can a reference to a precipitation method, which involves a completely wet liquid technique from which compounds settle out, suggest the claimed method involving contacting a solid with a solution only to the point of beginning wetness (impregnation to incipient wetness)? The rejection of Claim 28 must be removed.

In view of the above, it is submitted that all of Claims 10-14, 16-17, 19, 21, and 28 meet the requirements for non-obviousness. Accordingly, it is respectfully requested that the Final Rejection under 35 USC 103(a) be reversed.

C. Concerning the Final Rejection of Claims 10-17 Under 35 USC 103(a) in View of Haruta '394 taken with Hirose et al.

Claims 10-17 stand finally rejected under 35 USC 103(a) as allegedly being unpatentable over Haruta '394, taken as described hereinabove, in view of Hirose et. al., for the reasons of record. This rejection is respectfully appealed for the following reasons.

C.1. Arguments in Favor of Unobviousness of Subgroup C¹: Claims 10, 13-16

The Final Rejection attempts to validate Hirose et al. on the grounds that this citation is related to the catalyst arts and discloses a "combination of acetylacetonate of a metal ... with a reducing agent." The argument fails on two accounts. First, case law requires consideration of whether the prior art would have motivated or suggested the combination of references to those of ordinary skill in the art at the time the invention was made. *In re Gordon*, 733 F.2d 900, 221 USPQ 1125, 1127 (Fed. Cir. 1984). Appellant can find no motivation to combine Haruta '394 with Hirose et al. The former reference lies in the field of hydro-oxidation processes and oxidation catalysts; the latter reference lies in the field of polyolefin laminates, wherein the polyolefin is produced by a ring-opening polymerization reaction. There would be no motivation for a skilled artisan interested in improving a method of preparing gold-titanium hydro-oxidation catalysts to access art related to polyolefin laminates and ring-opening polymerization reactions. Second, it is to be recognized that the catalyst arts is a wide-ranging field and notoriously unpredictable. *In re Carleton*, 599 F.2d 1021; 202 U.S.P.Q. 165 (CCPA 1979); *In re Slocombe*, 510 F.2d 1398; 184 U.S.P.Q. 740 (CCPA, 1975); *In re Doumani et al.*, 281 F.2d 215 (CCPA, 1960); 126 USPQ 408. In consideration of this unpredictability, a citation must be close or pertinent to the field of the invention. Hirose et al. meets none of these criteria. Accordingly, Hirose et al. has no place in the instant rejection and must be removed.

Further to the point, Appellant even disagrees that Hirose et al. discloses what the Examiner says it does. By Appellant's reading, Hirose et al. teaches the titanyl acetylacetonate as being used in combination with "an organoaluminum

compound," which is not at all pertinent to the instant claims. Accordingly, Hirose et al. contributes nothing to the rejection. Claims 10 and 13-16 are unobvious over the combination of Haruta '394 and Hirose et al.

C.2. Additional Argument Favoring Unobviousness of Subgroup C²:

Claims 11 and 12

Claims 11 and 12 are drawn to reducing agents comprising organotitanium compounds, described in the specification as compounds containing Ti-C σ - or π -bonds, such as found in titanium cyclopentadienyl or alkyltitanium compounds. None of Haruta '394 or Hirose et al. teaches, suggests, or hints at the use of such compounds as reducing agents. See again Section B.2. above.

C.3. Additional Argument in Favor of Unobviousness of Subgroup C³: Claim 17

Claim 17 is drawn to a specific titanium loading. Hirose et al. adds nothing to the disclosure or suggestion of titanium loading. As to Haruta '394, refer to comments in Section B.3. hereinabove and calculations in Appendix B, attached hereto. Claim 17 is clearly unobvious over the combined references.

In view of the above, it is submitted that Claims 10-17 meet the requirements of unobviousness over the combined references to Haruta '394 and Hirose et al. Accordingly, it is respectfully requested that the Final Rejection of Claims 10-17 under 35 USC 103(a) be reversed.

D. Relevance of EP-A1-0,709,360

Appellant submitted into the record Haruta 'EP (EP-A1-0,709,360) as teaching a distinction between precipitation and impregnation techniques, and also as teaching away from impregnation methods for preparing catalysts comprising gold on titania, for use in hydro-oxidation processes. By teaching away from Appellant's claims method, Haruta 'EP supports Appellant's claims to unobviousness. The Examiner disposes of Haruta 'EP as "not relied upon" and as "not teaching away from impregnation." The Examiner makes a fatal mistake by ignoring a highly material

prior art reference.. All evidence of non-obviousness must be considered when assessing patentability. *In re Soni*, 34 USPQ2d, 1684, 1687 (Fed. Cir. 1995) The

First, it is crucial to understand that Haruta 'EP *distinguishes* between the deposition precipitation and impregnation methods in preparing a catalyst of gold on titania. The Board's attention is directed to Examples 1 and 3 of Haruta 'EP. Example 1 is drawn to preparation of a catalyst by deposition precipitation and Example 3 to its use thereof in oxidation of propylene with oxygen in the presence of hydrogen to prepare propylene oxide. An active catalyst is illustrated. By contrast, Comparative Example 1 in Haruta 'EP is drawn to a gold/titania catalyst prepared by impregnation and its use in the same process. Significantly, Haruta 'EP teaches that the catalyst prepared by impregnation *fails to produce* any of the desired propylene oxide product. *Hence, Haruta, himself, teaches to a distinction between deposition precipitation and impregnation methods of catalyst preparation and teaches away from impregnation for making active catalysts.* Secondly, it is crucial to understand that Haruta 'EP *does not teach, suggest, or hint at the use of an in situ reducing agent combined with the impregnation technique.* In contrast, Appellant has now discovered that when a *reducing agent is impregnated onto the support with gold impregnation*, then advantageously, the easiest method of preparing catalysts ---e.g., impregnation --- can be employed to prepare active gold-titanium catalysts useful for hydro-oxidation processes. Appellant's process achieves the desired olefin oxide in high selectivity (>90 mole percent) and at acceptable olefin conversions and desirably low H₂O/PO molar ratios (<10/1).

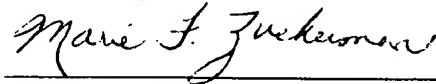
In view of the above, Appellant maintains that Haruta 'EP is a relevant reference that by manner of "teaching away" from aspects of the claimed invention, favors the patentability of the appealed claims.

E. CONCLUSION

In view of the arguments set forth herein, Appellant believes that appealed Claims 1-32 clearly meet all of the statutory requirements for patentability.

Accordingly, reversal of the Final Rejection is requested, and a favorable decision by the Board of Patent Appeals and Interferences is respectfully solicited.

Respectfully submitted,



Marie F. Zuckerman
Agent for A. Kuperman et al.
Registration No. 31,315
Phone: (203) 248-3907

Date: May 15, 2002

P.O. Box 1967
Midland, MI 48641-1967

Appendix A (Claims), attached hereto.
Appendix B (Calculations), attached hereto.

APPENDIX A

CLAIMS ON APPEAL

1. A process of preparing a catalyst composition comprising gold on a titanium-containing support, the process comprising impregnating a gold compound and a reducing agent onto a catalyst support, wherein the reducing agent comprises titanium, or the catalyst support comprises titanium, or both the reducing agent and the catalyst support comprise titanium, under conditions sufficient to prepare the catalyst composition.
2. The process of Claim 1 wherein the gold compound is selected from the group consisting of chloroauric acid, sodium chloroaurate, potassium chloroaurate, gold cyanide, potassium gold cyanide, diethylamine auric acid trichloride, gold acetate, alkyl gold halides, and alkali aurates.
3. The process of Claim 1 wherein the process is conducted at a gold loading greater than about 10 parts per million by weight, based on the total weight of the gold and support.
4. The process of Claim 1 wherein the reducing agent is an organic compound which does not contain titanium.
5. The process of Claim 4 wherein the reducing agent is selected from the group consisting of sugars, carboxylic acids and salts thereof, alcohols and alkoxide salts thereof, alkanolamines, alkylamines, and mixtures thereof.
6. The process of Claim 4 wherein the reducing agent is selected from the group consisting of C₆₋₂₀ sugars, C₂₋₂₀ carboxylic acids, C₁₋₁₅ aliphatic alcohols, C₁₋₁₅ alkylamines, the alkali and alkaline earth salts of the aforementioned sugars, carboxylic acids, and alcohols, and mixture of any of the aforementioned compounds.

7. The process of Claim 4 wherein the reducing agent is selected from the group consisting of methanol, ethanol, isopropanol, ethanolamine, acetic acid, lactic acid, citric acid, maleic acid, cinnamic acid, sodium acetate, sodium lactate, sodium citrate, sodium cinnamate, sodium maleate, and mixtures thereof.

8. The process of Claim 4 wherein the molar ratio of reducing agent to gold is greater than 0.5:1.

9. The process of Claim 4 wherein the organic reducing agent functions as a solvent for the impregnation.

10. The process of Claim 1 wherein the reducing agent contains titanium.

11. The process of Claim 10 wherein the reducing agent is an organotitanium compound.

12. The process of Claim 11 wherein the organotitanium compound is selected from the group consisting of alkyltitanium compounds and cyclopentadienyl titanium compounds.

13. The process of Claim 10 wherein the reducing agent is a titanium coordination compound.

14. The process of Claim 13 wherein the titanium coordination compound is selected from the group consisting of titanium alkoxides and titanium carboxylates.

15. The process of Claim 14 wherein the titanium coordination compound is titanyl acetylacetonate.

16. The process of Claim 10 wherein the gold compound is deposited onto the support prior to deposition of the titanium-containing reducing agent.

17. The process of Claim 10 wherein the process is conducted at a titanium loading of the titanium-containing reducing agent greater than 0.02 weight percent and less than 20 weight percent, based on the weight of the support.

18. The process of Claim 1 wherein the catalyst support is selected from the group consisting of titanium dioxide, titanosilicates, titanium dispersed on silica, promoter metal titanates, titanium dispersed on promoter metal silicates, and mixtures thereof.

19. The process of Claim 1 wherein the reducing agent contains titanium and the support is selected from the group consisting of silicas, aluminas, aluminosilicates, zirconia, magnesia, carbon, titania, and mixtures thereof.

20. The process of Claim 1 wherein the process is conducted at a titanium loading on the support of greater than 0.02 weight percent and less than 20 weight percent, based on the weight of the support.

21. The process of Claim 1 wherein both the reducing agent and the support contain titanium.

22. The process of Claim 1 wherein at least one promoter metal compound is impregnated onto the support.

23. The process of Claim 22 wherein the promoter metal is selected from the group consisting of silver, Group 1, Group 2, the lanthanide rare earth metals, the actinide metals of the Periodic Table, and mixtures thereof.

24. The process of Claim 22 wherein the total concentration of promoter metal or metals ranges from greater than about 0.01 to less than about 20 weight percent, based on the total weight of the catalyst.

25. The process of Claim 1 wherein the solvent for the impregnation is selected from the group consisting of water, organic solvents, and mixtures thereof.
26. The process of Claim 1 wherein after impregnation, the support is washed.
27. The process of Claim 26 wherein after washing, the support is treated with a solution containing at least one promoter metal.
28. The process of Claim 1 wherein the impregnation is conducted to the point of incipient wetness or a point of lesser wetness.
29. The process of Claim 1 wherein the impregnation is conducted at a temperature between 21°C and 100°C.
30. The process of Claim 1 wherein after impregnation and any additional steps of washing and treating with a promoter metal, the catalyst is heated.
31. The process of Claim 30 wherein the heating is conducted at a temperature greater than 250°C and less than 800°C.
32. The process of Claim 30 wherein the catalyst is heated in oxygen or an oxygen-containing gas, or heated in an inert atmosphere, or heated in a reducing atmosphere.

APPENDIX B1. CALCULATIONS OF PERCENTAGE TITANIUM, BY WEIGHT AS DISCLOSED BY HARUTA '394a. Weight percentage of Ti in TiO₂

$$\begin{array}{rcl}
 1 \times \text{Ti atomic weight (47.9 g/mole)} & = & 47.9 \\
 2 \times \text{O atomic weight (16.0 g/mole)} & = & 32.0 \\
 \text{Total Weight} & = & 79.9 \text{ g/ mole}
 \end{array}$$

$$\% \text{ Ti} = (47.9 / 79.9) \times 100\% = 59.9 \%$$

b. Weight percentage of Ti in Mixture of Au and TiO₂ (Au/Ti = 1/19)

$$\begin{array}{rcl}
 1 \times \text{Au atomic weight (197.0 g/mole)} & = & 197.0 \\
 19 \times 1 \times \text{Ti atomic weight (47.9 g/mole)} & = & 910.1 \\
 19 \times 2 \times \text{O atomic weight (16.0 g/mole)} & = & 608.0 \\
 \text{Total Weight} & = & 1715.1 \text{ g/mole}
 \end{array}$$

$$\% \text{ Ti} = (910.1 / 1715.1) \times 100\% = 53.1 \%$$

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2. Ti LOADING CLAIMED BY APPELLANT

0.02 to 20 weight percent, based on weight of support

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TC 1700

3. CONCLUSION

Appellant's claimed range lies outside the values inherently or implicitly disclosed by Haruta '394.



HF 11700
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

Applicant (s): Alex Kuperman et al.

Serial No.: 09/544,742

Group Art Unit: 1754

Filed: April 7, 2000

Examiner: Steven P. Griffin

For: METHOD OF PREPARING A CATALYST CONTAINING GOLD AND TITANIUM

COPY OF PAPERS
ORIGINALLY FILED

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Sir:

BRIEF FOR APPELLANT - FEE SHEET

This is an appeal to the Board of Appeals from the action of the Primary Examiner finally rejecting Claims 1-32, in the above-identified patent application.

Please charge the \$320.00 fee to our Deposit Account No. 04-1512. If this amount is incorrect, please charge or credit our account accordingly. One original and two duplicate copies of this sheet are enclosed.

Respectfully submitted,

Marie F. Zuckerman

Marie F. Zuckerman

Registration No. 31,315

Phone: 203-248-3907

P. O. Box 1967
Midland, MI 48641-1967